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Original Article

Dentistry Section

Quantitative Assessment of Fluoride Release and Recharge Ability of Different Restorative Materials in Different Media: An in Vitro Study

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ABSTRACT

Objective: To measure fluoride release and recharge ability of restorative materials in deionised water, artificial saliva and lactic acid.

Materials and Methods: Pellets were prepared from GC2, Ketac N100 and Beautifil II. Each pellets were individually immersed in 10 ml deionised water, artificial saliva or lactic acid as per respective subgroup for 24 h and then elutes were collected. Specimens were reimmersed in respective container. Fluoride released was analysed after 24 h, 7th and 15th day. On 15th day all specimens were exposed to 1.23% APF gel and fluoride release in respective solution was measured on 16th, 22nd, 30th day.

Result: Fluoride release was more after 24 h for all materials in all media then decrease gradually. GC2 shows more fluoride release than Ketac N100 at 24 hours and on 7th day but onwards Ketac N100 released significantly more fluoride. Beautifil II showed least fluoride release at all measured intervals in all media. Order of fluoride release in media was lactic acid > deionised water > artificial saliva for all materials.

Conclusion: GICs are smart material which release more fluoride when environment become more acidic and also show tendency to recharge which helps clinically in caries risk children.

Keywords: Artificial saliva, Fluoride release, Lactic acid, Recharge

INTRODUCTION

Glass ionomer cement is boon for pediatric and preventive dentistry as it is the adhesive material of choice to treat high-caries risk patients. Restorative material should not only restore the tooth but also help in prevention of caries by reducing caries risk [1]. Fluoride containing dental materials show clear differences in the fluoride release and uptake characteristics and may act as fluoride reservoir to increase fluoride level in saliva, plaque and hard dental tissues, or may help to prevent or reduce caries [2,3].

The fluoride release of glass ionomers depends on the type of glass ionomer, the initial fluoride content of the glass, mixing and setting times, and pH changes in the environment. It also reduces dental caries through promoting remineralization and influencing the morphology of teeth by reducing the solubility of enamel [4-8].

Considering the fact that topical fluoride can recharge exhausted glass ionomer cements, converting them into fluoride reservoirs allows them to constantly release fluoride. The present study has been conducted to evaluate the fluoride release and recharge (uptake) from different formulations of glass ionomer cements in deionised water, artificial saliva, and lactic acid.

MATERIALS AND METHODS

The materials used in this study , includes 3 glass ionomer cements conventional GIC (GC 2), Resin modified GIC (Ketac N100) and giomer (Beautifil II). The characteristics of used materials in study are given in [Table/Fig-1].

Sample Preparation

Specimens were prepared from 3 restorative materials. Sixty pellets were made from each material resulting in a total of 180 pellets. Pellets prepared from these restorative materials were mixed and / or cured as per manufacturer's instructions. Sufficient amount of material was placed into a brass mould with a specification of 5 mm x 2.5 mm which insured standardization of shape and size of each pellet. The material was pressed between mylar strips supported by glass slabs on either side. For the light cure materials (Beautifil II and Ketac N100) glass slab were replaced by glass slides during curing [9]. The pellets were allowed to set at room temperature for 15 min [10]. Finishing procedures were not incorporated as surface was cured against matrix strips which resulted in a satisfactory finish [11].

Restorative material	Composition	Manufacturer , Lot number and shade
Group 1- Conventional GIC	Powder: calcium fluoroalumino silicate glass, polyacrylic acid powder, iron oxide Liquid: polyacrylic acid (aqueous solution), tartaric acid , water	GC Gold label 2, GC corporation, Japan. Lot: 1104061 Shade -22 yellow brown
Group II- Resin Modified GIC	Paste A-fluoroalumino silicate glass, silane treated silica and zirconia silica nanofillers , methacrylate and dimethacrylate resin and photoinitiators Paste B-polyalkenoic acid copolymer, silane treated zirconia silica nanoclusters, silane-treated silica nanofillers and hydroxy- ethyl methacrylate	Ketac N100; 3M ESPE, St Paul MN, USA Lot: N266970 Shade: A2
Group III- Giomer	S-PRG glass filler, fluoride containing fluoro-boro-alumino silicate glass filler paticles, TEGDMA, Bis–GMA (17% weight)	Beautifil II; shofu Inc, Kyoto, Japan Lot: 41134 Shade-A2

[Table/Fig-1]: Restorative Materials used in the study

Division and Treatment of Specimens

Sixty pellets of each material prepared to make a total of 180 pellets were grouped and Each pellet was individually immersed in 10 ml deionised water [Table/Fig-2], artificial saliva or lactic acid as per respective sub group in an air tight container for 24 h.

Group	Sub Group	No. of specimens
I GC Gold label 2	IA – Deionised water	20
	IB – Artificial saliva	20
	IC – Lactic acid	20
II Ketac N100	IIA - Deionised water	20
	IIB - Artificial saliva	20
	IIIC – Lactic acid	20
III Beautifil II	IIIA - Deionised water	20
	IIIB - Artificial saliva	20
	IIIC – Lactic acid	20

[Table/Fig-2]: Division of samples

Collection of Elutes

After 24 h, the specimens were removed from the air tight container and the deionised water, artificial saliva and lactic acid elutes were collected. The specimens were then reimmersed in the air tight container containing 10 ml of fresh deionised water, artificial saliva and lactic acid as per the respective subgroup. The collection of elutes and refilling of fresh deionised water, artificial saliva and lactic acid was repeated every day. The amount of fluoride ions released in the solutions was analysed after 24 h, 7th day and 15th day.

Fluoride Recharge

After 15 days, initial fluoride release all specimen from each material were exposed to 1.23% APF gel (Bubble Gum Pascal (U.S.A.) for 4 minutes. After 4 min, excess gel was vigorously washed off for 30 sec with deionised water; the pellets were dried with absorbent paper and then reimmersed in the air tight container containing 10 ml of fresh deionised water, artificial saliva and lactic acid as per the respective subgroup. Fluoride analysis after application of APF gel was carried out on 16th, 22nd, 30th day.

Determination of Fluoride Release

To determine the amount of fluoride release, each 10 ml of elute was buffered with 1ml of total ionic strengths buffer (TISAB III). The fluoride concentration was measured with a specific fluoride ion electrode (Ino Lab pH/ION 735, WTW 82362 Wellheim, Germany) in part per million (ppm) by digital ion analyzer. The electrode was maintained according to the manufacturer's instructions and calibrated with standard solutions. The following concentrations of standard solutions: 0.1, 1, 10, 50 and 100 ppm were prepared from a 100 ppm F stock solution. After every 10 measurements the electrode was checked (recalibrated) with the standard solutions 1 and 10 ppm. The collected data was subjected to statistical analysis.

STATISTICAL ANALYSIS

The fluoride release (ppm) of various dental restorative materials in different media before and after application of APF gel was analyzed by using SPSS 17.0 .Mean and standard deviation for fluoride release values were obtained. Intergroup comparison was done by using Mann Whitney U-test and multiple intergroup comparisons by using Kruskal-Wallis Test probability (P) value $P < 0.05$ was considered to be statistically significant.

RESULTS

The values of fluoride release were tabulated as mean ± standard deviation. Result of current study showed in [Table/Fig-3-5]. All restorative materials in the study showed more release after 24 h, this was decreased rapidly on 7th day and 15th day in all the three media deionised water, artificial saliva and lactic acid. After APF gel application similar pattern was observed for all restorative material. More fluoride was released on 16th day and then decrease rapidly on 22nd and 30th day in all the three media.

In all media deionised water, artificial saliva and lactic acid fluoride release was maximum for conventional GIC (GC gold label 2- group IA) followed by resin modified GIC (Ketac N100- group IIA) and giomer (Beautifil II-group IIIA) respectively after 24 h and on 7thday. However, resin modified GIC (Ketac N100- group IIA) released more fluoride than conventional GIC (GC gold label 2-group IA) on 15th day. On 16th, 22nd and 30th day after application of APF gel, fluoride release was maximum for resin modified GIC (Ketac N100- group IIA) followed by conventional GIC (GC gold label 2-group IA) and giomer (Beautifil II- group IIIA) respectively. Difference in the amount of fluoride released in all three media for all restorative materials (group IA, IIA and IIIA) at all the measurement intervals was statistically significant ($p < 0.05$).

Fluoride release in lactic acid was more followed by deionised water followed by artificial saliva and this difference in fluoride release was statistically significant ($p < 0.05$).

The results of the present study are summarized as:

- Fluoride release by conventional GIC (GC Gold label 2), resin modified GIC (Ketac N100) and giomer (Beautifil II) at all measurement interval in all media was in following order- Lactic acid > Deionised water > Artificial saliva

	Before application of 1.23% APF Gel			After application of 1.23% APF Gel		
	After 24 hour	7th day	15th day	16th day	22nd day	30th day
GC 2	9.991 ± 0.456	1.521± 0.102	0.671± 0.024	9.907±0.531	1.324±0.126	0.545 ± 0.028
Ketac N100	6.014±0.222	1.037±0.075	0.753±0.020	11.880±0.503	1.994±0.127	1.082±0.098
Beautifil II	1.376±0.167	0.246±0.014	0.083±0.006	2.715±0.112	0.359±0.033	0.070±0.005

[Table/Fig-3]: Fluoride release in deionised water

	Before application of 1.23% APF Gel			After application of 1.23% APF Gel		
	After 24 hour	7th day	15th day	16th day	22nd day	30th day
GC 2	6.975 ±0.299	1.062 ± 0.089	0.552 ± 0.022	6.484 ± 0.311	0.997 ± 0.062	0.442 ± 0.023
Ketac N100	5.012± 0.166	0.903± 0.011	0.602± 0.018	8.818± 0.556	1.634±0.133	0.913±0.016
Beautifil II	1.024±0.140	0.172±0.013	0.072± 0.005	2.285±0.127	0.261±0.029	0.061±0.002

[Table/Fig-4]: Fluoride release in artificial saliva

	Before application of 1.23% APF Gel			After application of 1.23% APF Gel		
	After 24 hour	7th day	15th day	16th day	22nd day	30th day
GC 2	22.293±1.263	3.533±0.389	1.312±0.133	20.077±0.874	2.61±0.173	1.096±0.086
Ketac N100	16.088±0.692	2.825±0.099	1.552±0.133	22.125±1.376	4.044±0.161	2.076±0.170
Beautifil II	3.992±0.163	0.689±0.019	0.308±0.014	5.958±0.179	0.846±0.037	0.207±0.012

[Table/Fig-5]: Fluoride release in lactic acid

- Fluoride released by all the restorative material was maximum after 24 h then decreased on 7th day and 15th day. After application of APF gel fluoride release was maximum on 16th day and then decreased on 22nd and 30th day in all the media.
- All restorative material released fluoride in all media in following order.

After 24 hours and on 7th day- Conventional GIC (GC gold label 2) > Resin modified GIC (Ketac N100) > Giomer (Beautifil II).

On 15th day – Resin modified GIC (Ketac N100) > Conventional GIC (GC gold label 2) > Giomer (Beautifil II).

After Application of APF Gel

On 16th, 22nd and 30th day- Resin modified GIC (Ketac N100) > Conventional GIC (GC gold label 2) > Giomer (Beautifil II).

DISCUSSION

Glass ionomer cement releases fluoride into the surrounding tooth tissue immediately after setting and it is also capable of recharging itself. Since there is no existing ion in deionized water, the use of this is considered as giving an accurate estimate of fluoride released. This view is endorsed by authors [12-14]. Artificial saliva has been proposed in the present study in an attempt to offer more reliable oral conditions and it is also supported by other authors [2,3,15]. pH of the environment strongly affect the fluoride release from material. In early childhood caries cases pH of oral cavity decreases below critical pH and oral environment becomes acidic So, lactic acid (pH 5.2) was used to simulate critical pH and cariogenic conditions and it is supported by other investigators [16-18].

All restorative materials showed more release after 24 h, this was decreased on 7th day and 15th day. The fluoride release after 24 h was maximum because of surface wash off effect. Fluoride release from glass ionomer cement is diffusion limited and affected by concentration in both the cement matrix and the particles. During the initial acid dissolution of powder particle surfaces, a large amount of fluoride becomes part of reaction product matrix. This fluoride diffuse quickly from the matrix exposed on the surface of the material and is slowly replaced by fluoride diffusing from the matrix below the surface. This is responsible for the phenomenon of "burst effect", wherein high amount of fluoride are released after 24 h [19-21].

Fluoride release declines rapidly after 7th day then decreases gradually on 15th day. The probable explanation for this rapid decrease is release of fluoride occurs also by diffusion through pores and cracks. It is smaller but at a more constant level. This is presented by a long period of fluoride release at a nearly constant level 7-15 d after preparation of samples [22].

After APF gel application similar pattern was observed. More fluoride was released on 16th day and then decrease rapidly on 22nd and 30th day. This Initial high fluoride release followed by rapid decline after APF gel application as the fluoride can flow into the pores and cracks and be harbored there until the concentration of fluoride in the surrounding of the GIC is negative and the fluoride can be released again. This seems to be the process that is occurring after application of APF and the fluoride release which follows it [23].

The pH of deionised water causes surface loss of cement, whereas the high viscosity of artificial saliva retards the in and out diffusion of fluoride ions into the solutions [24]. It can be also be explained by the role of cations and anions present in the artificial saliva. These cations and anions react with the fluoride ions and retard the release from GICs. Hence, the values obtained in deionized water were noted higher than artificial saliva because the deionized water is free of any ions [24].

Also, our study showed that the fluoride release in deionised water was significantly less for all the tested restorative material when compared with fluoride release in lactic acid this results were in accordance with those of Kiran A et al., who evaluated a short term comparative analysis of Fluoride release from a newly introduced Glass Ionomer Cement in deionised water and lactic acid. Glass Ionomer Cements release more fluoride when the environment is at lower pH. This was supported by the observation that the amount of fluoride released was significantly higher throughout at pH 5.2 by many times greater than at neutral condition [25]. The significant difference in the amount of fluoride release from the materials in lactic acid and deionised water could be attributed to the fact that the dissolution of the material was dependent on the solvent.

Study demonstrated the order of fluoride release in all media before application of APF gel was After 24 hour and 7th day - GC gold label 2 > Ketac N100 > Beautifil II. 15th day – Ketac N100 > GC gold label 2 > Beautifil II.

Delbem AC et al., explained that the initial spike and sharp decline of fluoride release by GC gold label 2 can be attributed to its acid base setting reaction. Fluoride release occurs by two processes, first process correspond to the initial surface burst and second corresponds to long bulk diffusion. Conventional GIC set by an acid–base reaction between a degradable fluoride containing glass and a polyalkenoic acid. Initially all the fluoride is in the glass, but during the course of cement formation fluoride ions are released into the aqueous acid phase and become trapped in the hardening gel matrix. After setting and before the contact with water, the fluoride in the GIC is thus present in the remaining and not yet attacked leachable fluoride glass, associated to the polysalt matrix (complexed) and in the aqueous pore liquid (free) In the latter, the fluoride ions are only loosely bond and free to move. Fluoride ions are more available in conventional GIC. During the second process of fluoride release, diffusion of fluoride is higher in the glass–ionomer matrix. This may be due, to a more tightly bound and or less hydrophilic matrix of the resin modified GIC [19].

Wilson A D et al., [26] described the setting reaction of RMGICs as a dual setting one in which both acid- base and photo-polymerization take place. The acid- base reaction responsible for fluoride release as normal GICs and second reaction is a photo chemical polymerization process similar to light activated resin composite. Since, Ketac N100 sets by both acid-base as well as light polymerization reaction, it shows initial burst (1st-7th day) and afterwards (8th-15th day) gradual release of fluoride.

Difference in fluoride release of giomer (Beautifil II) and resin modified GIC is very well-explained as per Mousavinasab S et al., [1] who stated that initial setting of resin modified glass ionomer is performed by light activated polymerization followed by an acid base reaction arises from absorption of water. Beautifil II released less fluoride because it contains surface pre- reacted glass ionomer (S-PRG) as a fluoride component. The fluoride glass within Beautifil has little or no glass ionomers matrix phase, because of the lack of any significant acid base reaction. As fluoroalumino silicate glass and acid has been pre reacted water absorption is not critical in acid base reaction as is seen in this study.

In Our study fluoride release on 15th day was more by resin modified GIC (Ketac N100) than conventional GIC (GC gold label 2) this is explained by author [27] who found, a thinner hydrogel layer in resin modified GIC compared to thicker 300 nm, silica gel layer in conventional GIC that becomes thicker upon water sorption and can be cause for changing trend of fluoride release in resin modified gic (Ketac N100) on 15th day.

Our study demonstrated that fluoride release and recharge was maximum in lactic acid which simulates cariogenic condition. It means when condition becomes acidic due to cariogenic challenges GIC released more fluoride and act smartly by providing the greatest amount of fluoride when it would be most needed to prevent caries. Thus GIC behaves in a more dynamic fashion in the environment in which it is placed thus making it a smart material. GICs also shows both initial burst as well as long term release and also shows greater potential for recharge which help in prevention of caries.

CONCLUSION

Our study demonstrated that fluoride release and recharge was maximum in lactic acid which simulates cariogenic condition. It means when condition becomes acidic due to cariogenic challenges GIC released more fluoride whenever it required most. Thus GIC behaves in a more dynamic fashion in the environment in which it is placed thus making it a smart material. All glass ionomers used in the study showed both initial burst as well as long term release and also shows greater potential for recharge which help in prevention of caries.

This research was conducted in vitro, by considering only effect of media on fluoride release, whereas fluoride release may be modified by variables that are presented in vivo. So it is important to develop more in vivo studies with large sample size to assess different variables which influence physical and chemical behaviour of restorative materials intra orally. To take into account the dynamic factors present in oral cavity, further clinical studies combining both qualitative and quantitative evaluation are necessary.

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